Optical Refrigeration in the Solid State
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Motivation

Operating at reduced temperature dramatically enhances the performance of many devices. Semiconductor laser diodes are more efficient and semiconductor photodetectors are more sensitive when cooled. Some devices, particularly those that depend on the phenomenon of superconductivity, only operate at cryogenic temperatures (less than approximately 150 K). Hence, the need for reliable refrigeration of electronic devices is well established. However, existing refrigeration technology is far from ideal. Vibrations produced by Sterling cycle refrigerators are detrimental to device performance. Magnetic salt cooling is not compatible with many applications. Cryogenic baths require a continuous supply of cryogenic liquids, which are difficult to make and transport. And the minimum temperature achievable via Peltier cooling is only about 220K. Clearly, there is a need for a quiet, robust refrigerator that can achieve and maintain cryogenic temperatures.

The Basic Concept

We are investigating a new solid-state refrigeration scheme that addresses all of the devices needs described above. The mechanism relies on a unique optical absorption and emission process that extracts energy from a semiconductor. Some of this light is absorbed, creating cold carriers at the bottom of their respective bands. These carriers quickly thermalize with the semiconductor lattice, absorbing approximately $(3/2)kT$ of energy. Eventually, the carriers recombine to emit higher energy light than the light that was originally absorbed. In this way, heat is removed from the semiconductor.

This approach has several distinct advantages. First of all, there are no moving parts. If excitation is provided by semiconductor laser diodes, the refrigerator is completely solid state. Thus, it is expected to be very reliable. Secondly, the only external requirement is a current source to drive the laser diodes. The third advantage applies to the refrigeration of optical devices, which constitute the majority of semiconductor devices that perform better at cold temperatures. In this case, the optical nature of the cooling mechanism will greatly facilitate integration of the refrigerator and the device to be cooled.

Early Work on the Grant

In practice, the cooling mechanism that we are studying is not perfect. It is limited by the possibility of nonradiative recombination in the semiconductor. While each radiative event event contributes the entire bandgap energy to the system, giving rise to counterproductive heating. Since the bandgap energy is 35 times larger than $(3/2)kT$ at room temperature, the radiative efficiency must be very good to achieve net cooling.

This problem is aggravated by the high refractive index of the semiconductor material and the resulting difficulty of coupling photoluminescence out of the device. Without special index-matching efforts, the majority of the emitted light is totally internally reflected. This means

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that most of the photoluminescence is reabsorbed and re-emitted several times, with each cycle providing an additional opportunity for nonradiative recombination to occur. The limited escape cone of emitted light further restricts the internal radiative efficiency requirements of the semiconductor described above.

For approximately the first half of the three-year grant period, we concentrated our efforts on work with high purity GaAs. We used epitaxial liftoff to remove a very high-quality GaAs heterostructure from its absorbing substrate, then optically contacted the sample to a hemispherical ZnSe index-matching lens. Using this scheme, we observed an external emission efficiency of 96%, by far the highest ever observed from a semiconductor [1]. Compared to the previous record of 72%, the generation of heat was reduced a factor of 7. In fact, the device was only 1% shy of the efficiency required for net refrigeration.

Recent directions

Despite the early promise of GaAs as an active material, further progress was problematic. The epitaxial liftoff procedure was not very reproducible, and the optical contacting step often resulted in unacceptable damage to the sample. Tiny cracks running through the sample were loci for nonradiative recombination. Our inability to develop a *reproducible* sample handling protocol made it impossible for us to make quantitative comparisons of differing crystal growth recipes.

Faced with these difficulties, we decided therefore to move to a very different active material. We realized that both the liftoff and the optical contacting procedure can be avoided if the heterostructure is grown on a wafer which is very transparent and also so thick that it can itself be formed into the index-matching lens. Accordingly, we began to work with InGaAs passivated with InP. The heterostructure was grown on a thick InP wafer, which was then polished into a parabolic shape to provide optimum outcoupling of florescent light [2]. The InGaAs bandgap energy is only half that of GaAs, which means that the heating due to a given nonradiative recombination event is only half as large. The primary shortcoming of the InGaAs/InP system is fast Auger scattering, a process that converts excitation into heat rather than light. We intended to circumvent this problem by operating at cryogenic temperatures, where the Auger rate becomes negligible.

Recent results.

Room temperature results on the InGaAs/InP system were encouraging. We obtained a radiative efficiency of 60%, far in excess of what is usually observed in a small-band-gap material. Using standard models for extending the radiative and nonradiative rates to low temperatures, our calculations suggested that our first sample would already be a break-even efficiency for cooling at 77 K [3].

The actual behavior observed in our optical-access cryostat at 77 K was quite disappointing. External radiative efficiency was not dramatically better at cryogenic temperatures than it had been at room temperature. The reason, in retrospect, is fairly obvious, but we did not anticipate it prior to actually doing the experiments. The dominant sources of nonradiative recombination are surface recombination and auger processes. The two processes scale as n and as n^3, respectively, where n is the free carrier density. The desirable radiative recombination rate scales as n^2. The optimum radiative efficiency is achieved

at a value of n for which the surface recombination and the auger processes contribute about equally to the nonradiative recombination. Going to cryogenic temperatures dramatically suppresses the auger rate, but to realize corresponding gains in radiative efficiency, one is required to operate at much higher carrier density, so as to increase the n-square radiative rate relative to the n-linear surface-recombination rate. Unfortunately, operating at lower density also has the effect of reducing the value of n for which carriers just at the band-edge form a saturated Fermi gas. These two facts work against one another, so that long before we get to the optimum value for n, the sample begins to lase. Lasing is fine as far as it goes, but (in contrast to simple florescence) it is not possible to arrange for laser light to be blue-shifted with respect to the pump light, so lasing can not result in cooling.

This problem of lasing at sub-optimum carrier density can be surmounted in principle by judicious doping of the sample, but adding dopants to a sample while maintaining near-perfect radiative properties is a major development project. We had barely begun to explore the parameters of the problem with our crystal-growing collaborator when our grant ran out, and we decided to wrap up the project.

Our work resulted in an interesting technological spin-off: for technical reasons, it was simpler for us to polish the substrate into a parabolic, rather than a hemispherical shape. The emitted florescence then preferentially came out in a collimated beam rather than isotropically. We discovered by accident that the direction of the isotropic emission depends sensitively on the exact location at which the excitation takes place. In view of the fact that this steerable beam of light is generated with record-high efficiency, a postdoc in our group is filing a patent application for a high-efficiency, steerable light source [4].

Outlook for Optical Cooling in the Solidstate

We have been unable to realize cooling with our optical scheme. We feel the basic idea is sound enough, and that the ultimate success of this concept hinges entirely on issues of materials. Working with two different semiconductor materials, we have been able to realize external radiative efficiencies superior to the previous state-of-the-art, but nonetheless inadequate for actual cooling. Progress beyond this point will require a very focused research effort into materials development and characterization, an effort which we feel is beyond the scope of our small university-based group. We note with interest and some pride that a local aerospace corporation, Ball, is aggressively pursuing optical refrigeration technology.

Patents, Inventions and Industrial Implications

The research funded by the ONR grant resulted …
(i) in one invention for which a patent that has already issued [5], on optical cooling in solids;
(ii) in a second invention for which a patent application is being prepared [4], for a directional, high-efficiency, light-emitting diode;
(3) in informal ongoing conversations with engineers at Ball Aerospace in Boulder. Ball is attempting to develop and commercialize optical refrigeration technology.

References

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[2] T. H. Gfroerer, A. E. Cornell, and M. W. Wanlass, "Efficient directional spontaneous emission from an InGaAs/InP heterostructure with an integral parabolic reflector," J. Appl. Phys. 84, 5360 (1998).

[3] T. H. Gfroerer and E. A. Cornell, "Progress towards laser cooling in semiconductors" in OSA TOPS Vol. 18, Radiative Processes and Dephasing in Semiconductors, D. S. Citrin (ed.) p. 39 (1998).

[4] T. H. Gfroerer, "Adjustable Beam Light Emitting (ABLE) Diode," invention disclosure. (1998).

[5] E. A. Cornell and M. J. Renn, "Optical Cooling of Solids," US Patent number 5,615,558 (1997).